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Influence of light distribution on the performance of photocatalytic reactors: LED vs mercury lamps

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ABSTRACT

UV LED technology has revolutionized the photocatalytic processes due to their significant advantages over traditional mercury-based illumination sources, specifically its higher energy efficiency. However, the use of LED also introduces important changes in the light distribution achieved inside a photocatalytic reactor that has to be considered. In this study an exhaustive comparison of three different UV-A sources (a mercury fluorescent lamp, a 8-LED based system and a 40-LED based system) with different light distribution has been carried out. Theoretical distribution of the light was modeled in Ansys Fluent v14.5. The results of photocatalytic activity for methanol oxidation show that a homogeneous light distribution allows achieving a higher photonic efficiency. The diffuse and uniform emission of the fluorescent mercury lamp partially compensates its lower energy efficiency, leading to similar results than the 8-LED system. This fact can be explained taking in consideration that electron-hole recombination is enhanced in the areas with higher radiation intensities, decreasing the overall efficiency. In the case of the 40 LED due to the improvement of the light homogeneity and energy efficiency, higher reaction rates per kWh were achieved. These results show that despite the advantages of LED, if light distribution is not optimized it can result in lighting systems less effective than traditional ones. On the opposite, for bacterial inactivation the results show that there is no clear difference when using different lighting sources. The existence of a highly non-uniform radiation field with regions of the reactor with very high intensities seems to enhance the efficiency of the direct bacterial inactivation when LED are used, compensating the decrease in the charge transfer efficiency of the semiconductor based photocatalytic process.

1. Introduction

The chlorination processes for disinfection of drinking water supplies have raised public concerns as a consequence of the formation of potentially harmful chloro-organic disinfection by-products (DBPs) coming from the reaction with naturally-occurring organic matter [1]. Among the processes currently in development, semiconductor photocatalysis has emerged as a very attractive and environmentally friendly technology for water disinfection [2,3]. TiO_2 has widely been used as photocatalyst due to its low cost and non-toxicity [4,5]. Because of its absorption spectrum, activation of TiO_2 requires a light source with a wavelength below 400 nm in the UV-A range. Conventional UV lamps such as low-pressure mercury lamps have been widely used [6]. However, these lamps provide a low conversion of the electrical energy input into useful light being the high consumption of energy one of the main drawbacks of the photocatalytic process [7,8]. Moreover these type of lamps contain trace amounts of toxic mercury, are fragile and rigid cylindrical, and have a relatively short life span (<12,000 h) [9,10]. The recent availability in the market of UV LED has produced a revolution in the field with an exponentially growing number of publications reporting photocatalytic applications based on LED light sources. They offer several advantages over classical incandescent and conventional mercury discharge lamps, offering a higher energy efficiency, a longer lifetime (5 times that of Hg-vapor lamps), easily adaptable output, instant on-off or tunable wavelength [11–13]. Moreover, the use of LED makes the reactor design significantly more flexible, not being constrained by the tubular shape of mercury lamps. However, the use of LED also introduces important changes in the light distribution achieved inside the reactor that must be taken into account. Few studies have been found in bibliography comparing the performance of the use UV-A LED and Hg-vapor UV-A lamps. Ghosh et al. [14] evaluated the efficacies for the photocatalytic oxidation of 4-chlorophenol using LED sources, conventional Hg lamps and sunlight [14]. The results demonstrated that high intensity LED performance was superior to sunlight, and similar to a conventional

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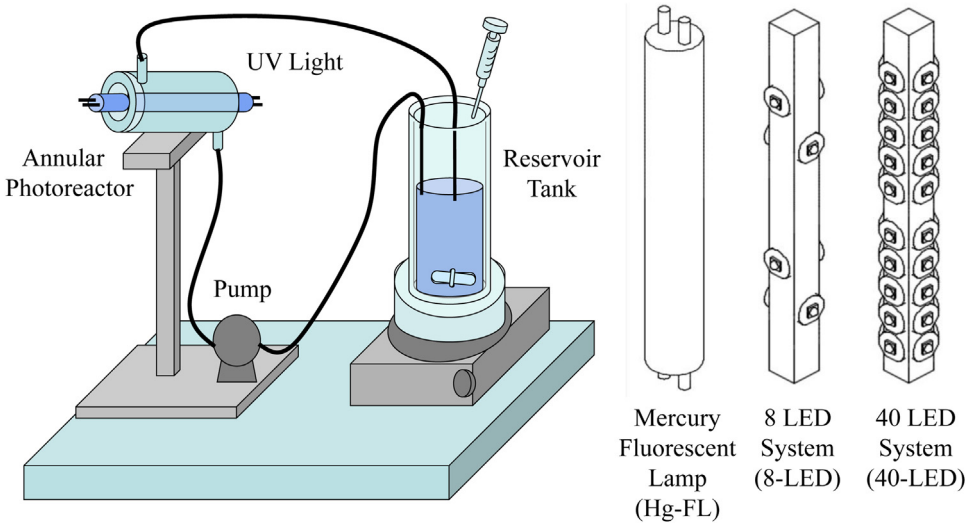

Fig. 1. a) Schematic representation of the experimental setup. b) Illumination sources.

lamp for the photocatalytic oxidation. Another study is the one carried out by Levine et al. [15] in which the efficiency of a LED based system and a UV-A fluorescent black light working at the same irradiance values for ethanol oxidation was compared. The results showed a lower removal efficiency when using LED. This behavior was attributed to a non-uniform irradiance over the photocatalyst. Similar results were obtained by Kim et al. in the photo-oxidation of cyanide [16]. Thus, a key question is how to select the optimal combination of variables related to light sources and reactor design simultaneously [17].

In this study, a novel approach has been applied for the exhaustive comparison of two different UV-A sources with different light distribution: a mercury fluorescent lamp (Hg-FL) and an 8 LED based system (8-LED). The study has been based on the theoretical calculation of the light distribution inside the reactor and its influence on the photocatalytic efficiency. Conclusions from this study led to the design of a third UV-A source based on a 40 LED system (40-LED) that allowed the validation of the hypothesis about how the distribution of light within the reactor influences the photonic efficiency. Theoretical distribution of the light was modeled to explain the relationship between the reaction rate and light distribution. In order to evaluate the photonic efficiency of each system and taking into account that several authors have pointed out the existence of certain differences between the photocatalytic oxidation of organics and the inactivation of microorganisms [18,19] two pollutants were chosen as models. The photocatalytic oxidation of methanol to formaldehyde was used as model chemical reaction, whereas *E. coli* was selected as model pathogenic microorganism for disinfection applications.

2. Experimental

2.1. Photocatalytic reactor

The photoreactor used to calculate the kinetic parameters, represented in Fig. 1**a**, consists of an annular photoreactor (15 cm long, 3 cm inner diameter and 5 cm outer diameter) operating in a closed recirculating circuit driven with a reservoir tank, being the total working volume of 1 L. As illumination source, a Philips TL 6W black light lamp and two LED-based systems with 8 and 40 LED (LedEngin Model LZ1-00UV00) were used (Fig. 1**b**), the three of them with a maximum emission peak centered at 365 nm (Fig. 2) and the same location in the axis of the annular photoreactor. The LED-based systems were continuously refrigerated using a liquid cooling system (Koolance EX2-755). The irradiation power of the Hg-FL was controlled using neutral filters with different opacity percentages as previously reported [20]. In the case of the LED systems, the irradiation power was controlled through the electrical current intensity using the software Eldoled LED driver configuration Toolbox. The total irradiation power in each case was calculated from potassium ferrioxalate actinometry experiments as described elsewhere [21].

2.2. Photocatalytic experiments

Photocatalytic experiments were carried out using commercial Evonik P25 titanium dioxide as photocatalyst at a concentration value of 0.1 g L^{-1}, previously optimized [22].

Methanol (Sigma-Aldrich, LC–MS) was chosen as model chemical pollutant at initial concentration of 100 mM. All solutions were done in deionized water. The oxidation of methanol was followed through the colorimetric determination of the formaldehyde produced throughout the reaction, quantitative oxidation product when methanol is in excess [23].

Escherichia coli K12 strains (CECT 4624, corresponding to ATCC 23631, where CECT stands for "Colección Española de Cultivos Tipo") were used to prepare the bacterial suspensions (NaCl 0.9%).

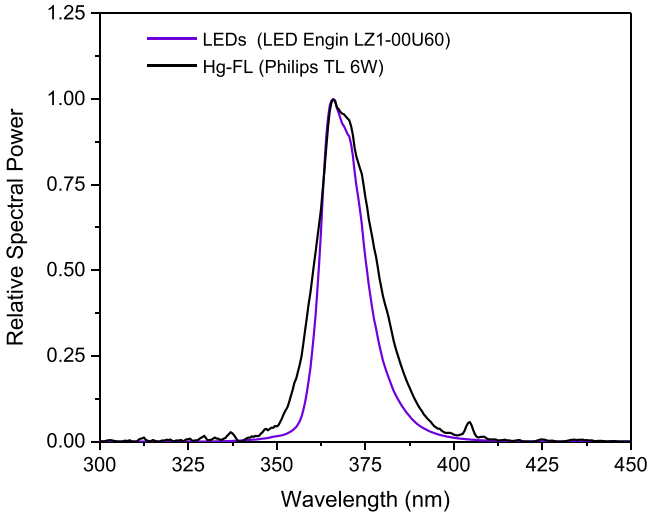

Fig. 2. Spectral distribution of the illumination sources.

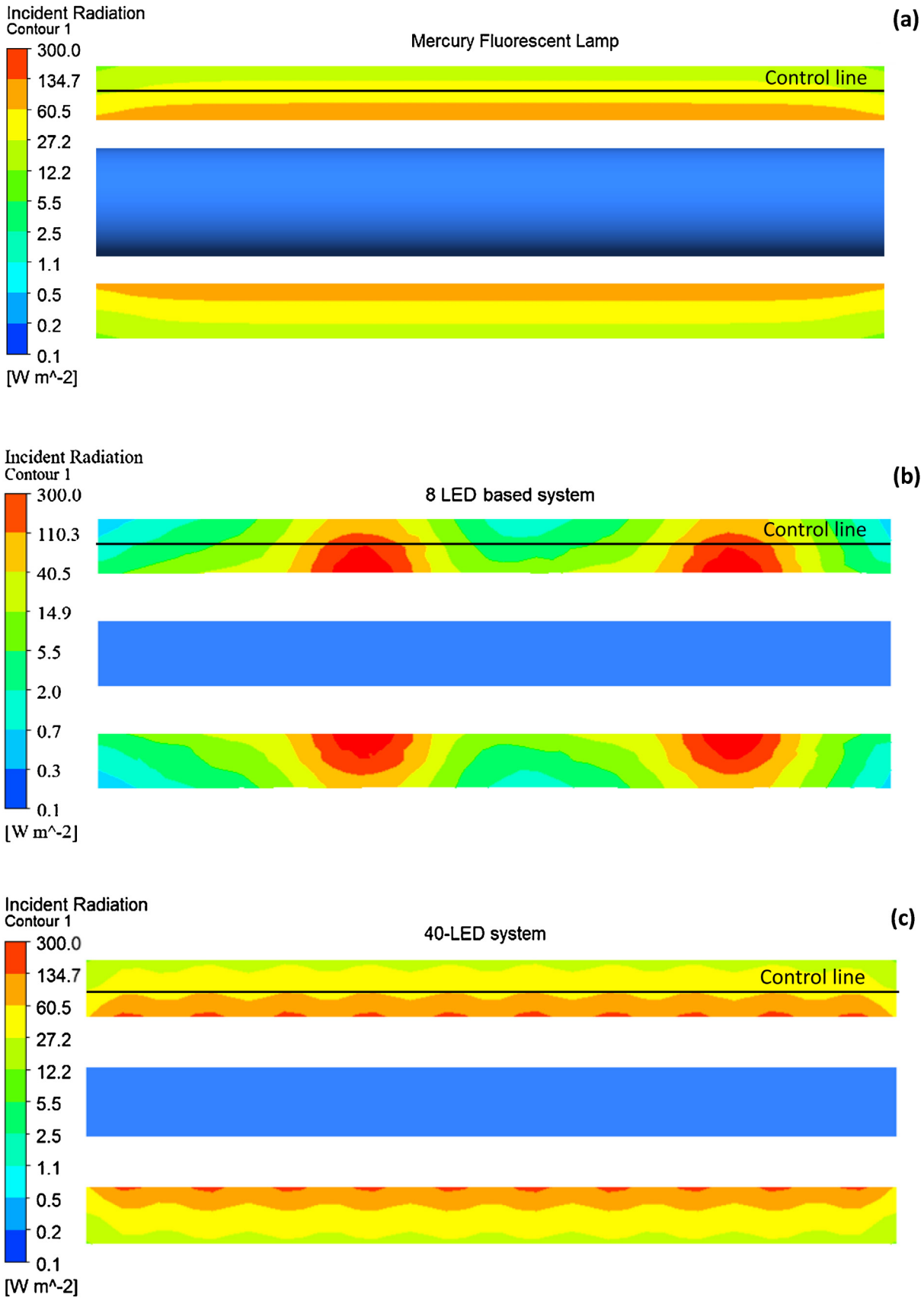

Fig. 3. Contours of incident radiation among a longitudinal plane inside the reactor when using (a) Hg-FL, (b) 8-LED, and (c) 40-LED for the same total emission.

Fresh liquid cultures were prepared by inoculation in a Luria-Bertani (LB) nutrient medium (Miller's LB Broth, Scharlab) and incubation at 37 °C for 24 h under constant stirring on a rotary shaker. An initial bacterial concentration of 10^6 CFU mL^{-1} was used in all the experiments. The analysis of the samples throughout the reaction was carried out following a standard serial dilution procedure. Each decimal dilution was spotted 4 times on LB nutrient agar plates and incubated at 37 °C for 24 h before counting. In order to test the reproducibility, all the experiments were performed at least twice being the results expressed as the mean and the experimental errors as the standard deviation.

2.3. Modelling of the photoreactor radiation field

The light distribution along the reactor with the different illumination sources was calculated using the commercial software Ansys 14.5 (Ansys Inc.®). The same dimensions of the experimental annular reactor were implemented for the calculation using the Ansys Workbench tool. The radiative transfer equation was solved using the discrete ordinate method obtaining the incident radiation at any point inside the reactor space. The optical properties of P25 TiO_2 aqueous suspensions were taken from the literature [24] and introduced into the model in order to consider absorption and scattering phenomena. The lamp and the LED surfaces were set as semi-transparent purely diffuse walls and the total external irradiance was fixed taking into account the actinometrical experiments. All the remaining walls were set as semi-transparent and also zero-thickness [25].

3. Results and discussion

3.1. Light distribution calculation

To choose the source of light to be used in a photocatalytic system it is necessary to consider the way in which the light is distributed throughout the reactor since this fact could play a very important role in the photonic efficiency of the system.

The incident radiation inside the reactor for each of the illumination sources was calculated with Ansys Fluent 14.5 software using the model described by Casado et al. [25]. In Fig. 3, the contours of incident radiation along a longitudinal plane within the reaction zone are shown for each illumination device when the same total emission is fixed. It is observed a significant disparity in terms of light distribution when different illumination sources are used.

The distribution of the incident radiation along of the control line showed in Fig. 3. for a total incident radiation value of 2.77×10^{-6} E/L s was also calculated and represented in Fig. 4. It can be observed how in the case of the Hg-FL, a very homogeneous distribution of the light is achieved whereas in the case of the 8-LED, there are highly irradiated areas corresponding to the place where the LED are located whereas other areas remain practically in darkness. On the other hand, the 40-LED system represents an intermediate case in which the homogeneity of light distribution has been improved with the increase of the number of LED but has not been able to reach the level of homogeneity achieved with the Hg-FL. It is necessary to note that the line studied is aligned with the LED array so it does not represent the totality of the reactor.

In order to have an exhaustive evaluation tool of light distribution along the reactor, the uniformity index of the incident radiation was calculated. This index represents how a variable field varies over a surface, where a value of 1 indicates the highest uniformity [26]. Five longitudinal planes were chosen to represent correctly the behavior of the whole reactor and the uniformity index was calculated on them. A weighted average of these values was made considering the total incident radiation in each plane. Incident radiation uniformity indexes of 0.685, 0.382, and 0.559 were obtained for the Hg-FL, the 8-LED system, and the 40-LED system respectively. These results confirm that the best distribution of light is obtained when the Hg-FL is used. It is also important to note that an increase in the number of LED leads to a significant increase in homogeneity of the reactor, which may have important implications in the photonic efficiency of the system. Uniformity index values were also calculated for different incident radiation values (data not shown). The results shown are similar to the previously obtained which means that the value of the total incident radiation does not have a significant effect on the homogeneity of the reactor when the same lighting source is used.

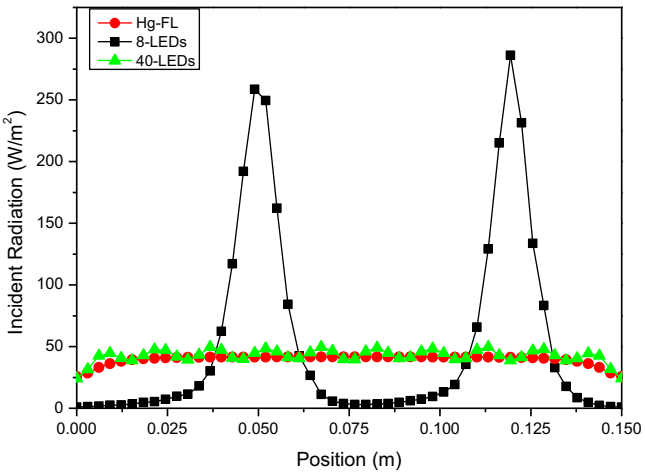

Fig. 4. Incident radiation calculated along a control line placed in the reactor area for a total incident radiation of 2.77×10^{-6} E/L s.

Table 1
Uniformity index for three different TiO_2 concentrations.

	Uniformity Index		
TiO_2 (g/L)	0.1	0.25	0.5
Hg-FL	0.685	0.579	0.493
8-LED	0.382	0.275	0.225
40-LED	0.559	0.477	0.424

Moreover, the uniformity index was calculated for higher values of concentration of TiO_2. The general trend observed was the decrease in the values of the uniformity index (Table 1) when the titania loading increases. It is likely due to a higher photon absorption in the areas closer to the light source, which causes a more noticeable decrease of incident light in the radial profile of the reactor.

3.2. Incident radiation and power consumption

Actinometrical experiments were carried out to determine the total amount of photons incident to the reactor volume for different emission conditions of the light sources. In the LED systems, the electric power was controlled in order to modify the incident radiation. In the case of the Hg-FL, neutral filters with different opacity percentage were used to modify the incident radiation but the electric power consumption remained the same.

As it is well known, an increase in the electrical current of LED causes an increase in the amount of emitted light. However, it is important to note that this relation is not completely linear, due to a decrease in the LED efficiency for higher current intensities. That means that the incident radiation is not proportional to the electric power consumption when high working current is used as confirmed by the results depicted in Fig. 5. For the same electric

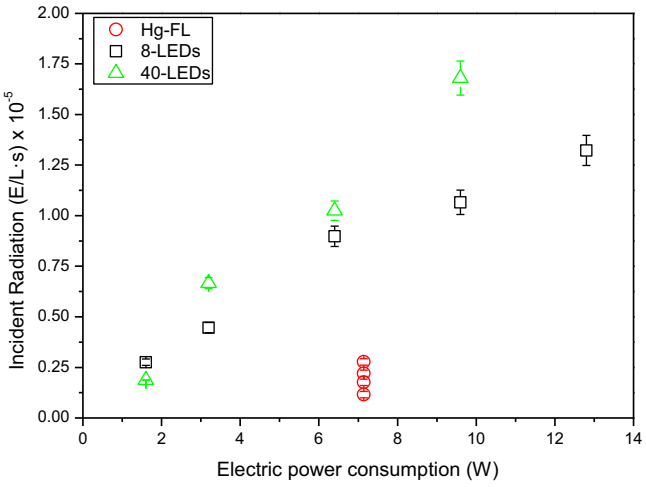

Fig. 5. Incident radiation versus power consumption for the three light sources.

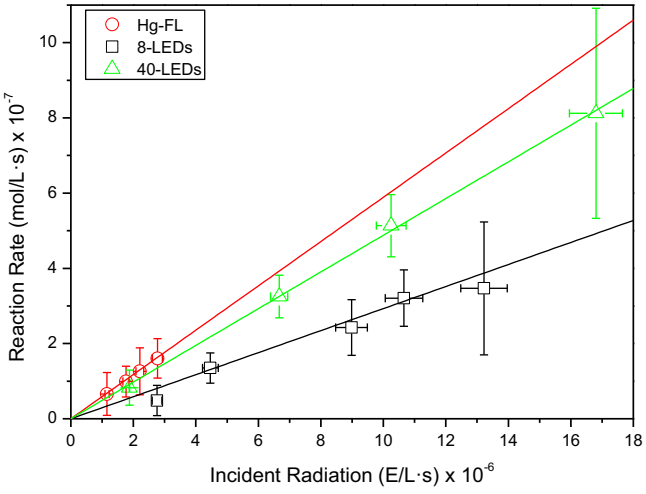

Fig. 6. Reaction rate of formaldehyde formation versus incident radiation for different illumination sources.

power consumption, the 40-LED system work with five times lower intensity since it presents a number of LED five times higher. If the efficiency of both systems in terms of photons produced per unit of energy consumed is calculated, it is obtained a mean value of 4.32 E/kW h for the 8-LED system and a value of 5.88 E/kW h for the 40-LED system. It can be concluded that when working under the current intervals studied a great improvement in the conversion of electricity to light is achieved for the 40-LED system. This result is very important for the overall efficiency of the global process. For the Hg-FL a value of 1.4 E/kW h is obtained when no neutral filter is used, being the system with the lowest electricity to light conversion efficiency, as expected.

3.3. Photocatalytic oxidation of methanol

The reaction rate of formaldehyde formation using the different light sources and power conditions are shown in Fig. 6. In all cases, a linear dependence between the reaction rate and the incident radiation is clearly observed. This behavior can easily be explained taking into account that, in this intensity range, an increase in the incident radiation means an increase in the energy supplied to the catalyst. This results in a proportionally larger generation of electron-hole pairs and therefore of hydroxyl radicals, leading to a linear increase in the reaction rate. On the other hand, this straight

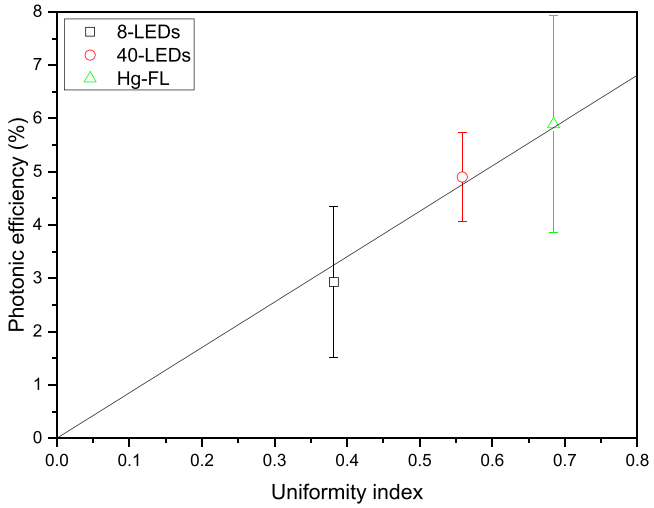

Fig. 7. Photonic efficiency versus the uniformity index calculated for each lighting system.

dependence means that the use of lower intensities will result in a proportionally linear decrease in the reaction rate, allowing an easy tune of the process with the regulation of the electrical intensity powering the LEDs. The results also seem to indicate that the values of reaction rate in the case of the Hg-FL are higher when normalized to the values of incident radiation, although this conclusion should be taken carefully considering the experimental uncertainty of the data derived of this extrapolation. A photonic efficiency of 5.89 is calculated for the Hg-FL whereas values of 3.5 and 4.9 are obtained for the 8-LED and 40-LED systems, respectively.

To explain this behavior it is necessary to focus on the distribution of light commented previously. In a study conducted by Boyjoo et al. [27] a numerical model of a reactor using a fluorescent lamp was developed. They determined that in the areas of the reactor closer to the lamp where the highest incident radiation is reached, the oxidation of ethanol follows a half order whereas in the regions where the incident radiation is lower the order of reaction is 1 [27].

As demonstrated by the calculation of the uniformity index, the uniformity of the light distribution along the reactor varies depending on the light source being used. In Figs. 3 and 4 it was observed that when using LED systems there are a greater proportion of reaction areas with high incident radiation concentrations than when the Hg-FL is used. According to the theory formulated by Boyjoo et al. [27] this means that when using LED greater areas of the reactor working following a half reaction order which consequently decreases the photonic efficiency of the system.

This behavior was also confirmed by Casado et al. [25]. They also demonstrated using a numerical simulation model that when working at higher incident radiation concentrations, a lower photonic efficiency is achieved due to the increase in the recombination effect of the electron-hole pairs.

Additionally, it is important to note the difference between the photonic efficiency values obtained for the two LED-based systems. It can be seen that an increase in the number of LED improves the photonic efficiency due to a greater homogeneity of the light distribution, being reduced the regions with noticeable high values of incident radiation. Fig. 7 confirms the existence of a direct relationship between the uniformity index calculated for each system and its corresponding photonic efficiency, which will determine the overall efficiency of the process.

When the results are analyzed in terms of the energy consumption, Fig. 8 shows that as a consequence of the great difference in energy efficiency between the Hg-FL and the two LED-based systems (Fig. 5), the difference in photonic efficiency is partially offset.

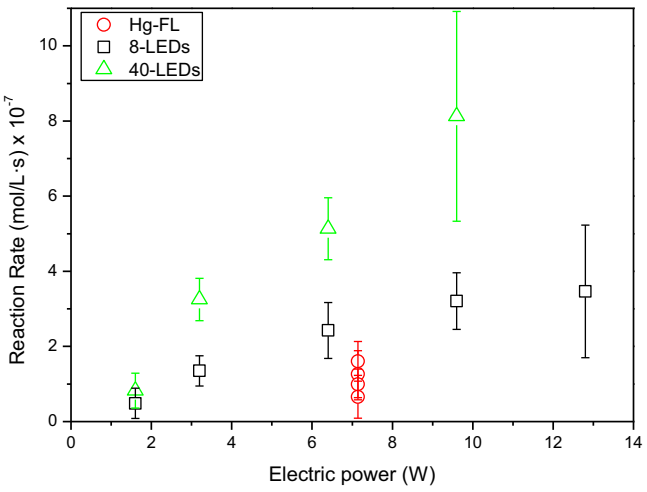

Fig. 8. Reaction rate of formaldehyde formation versus power consumption for different illumination sources.

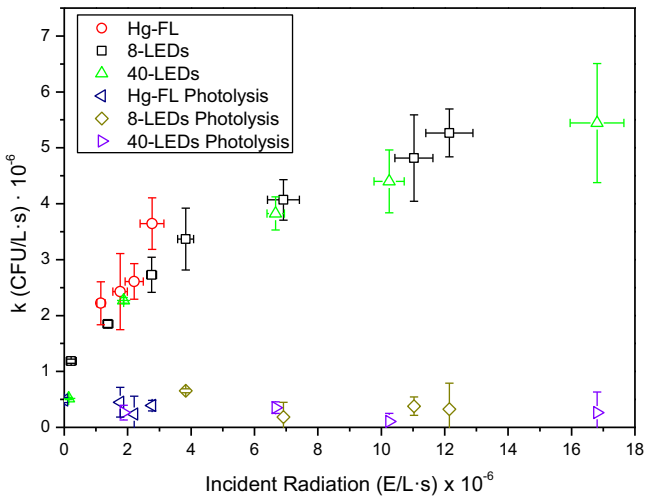

Fig. 9. Kinetic constants calculated for *E. coli* photocatalytic inactivation versus incident irradiation with the different light sources.

The 8-LED system is able to produce 0.166 mol of formaldehyde per kWh consumed. A value of 0.11 mol/kW h is achieved when the Hg-FL is used. The 40-LED is the most efficient system as it leads to a ratio of 0.367 mol/kW h due to the improvement of the light distribution compared to that of 8-LED, and the highest effectivity in the electricity conversion to light compared to that of Hg-FL.

These results demonstrate the critical role of light distribution in the efficiency of photocatalytic processes. Although in recent years the use of LED in photoreactor design has been widely spread based on its higher energy efficiency, our results confirm that if adequate reactor design is not carried out, the overall efficiency of the process could be not improved with respect to traditional mercury lamps.

3.4. Bacterial inactivation

Bacterial inactivation experiments under similar illumination conditions to those previously used in the methanol oxidation were carried out. The inactivation profiles were fitted using a mechanistic model previously reported [19] and based on a series-event mechanism for the cumulative attached of reactive oxygen species to the viable bacteria until their complete inactivation. The kinetic constant of this model, *k*, represents the reaction rate of the process, allowing the comparison of the efficiency of different experiments. Fig. 9 shows the kinetic constants with and without catalyst (photolysis) obtained for each illumination source when different values of incident radiation are used. Unlike what it was observed for methanol oxidation, there is no clear difference between the three illumination systems for bacterial inactivation.

These results are in apparent contradictions with previous conclusions derived of the methanol oxidation photocatalytic tests, where a more homogeneous distribution of light involves a higher photonic efficiency. A possible explanation for this behavior can be found in the work of Soomer et al. [28] where the inactivation of *E. coli* working with different irradiation values was studied. The dose was calculated as the total amount of incident radiation over the entire reaction time. It was observed that for the same system, when working with higher currents values for the same total dose of irradiation, greater values of bacterial inactivation were achieved. These results indicate that a higher effectiveness is reached when applying a high UV intensity for a short period of time in comparison with extending the same dose over a longer period of exposure. They attributed it to repair enzymes of the cell which are more influenced by high UV intensities. Similar conclusions were reached by Rincón and Pulgarin [29].

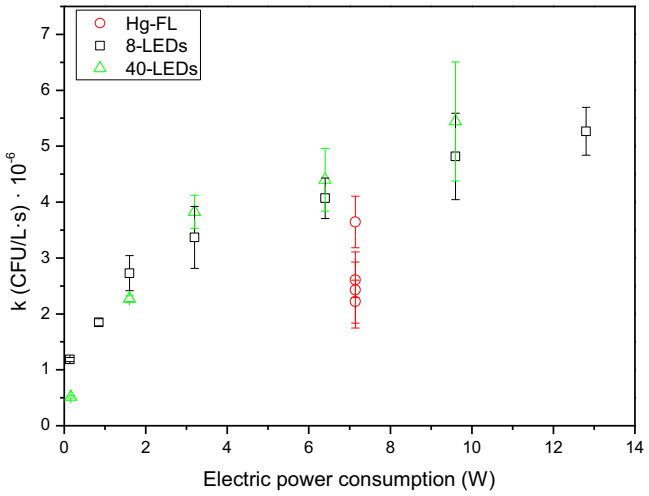

Fig. 10. Kinetic constants calculated for *E. coli* inactivation versus the power consumption for the different light sources.

Consequently, the results in Fig. 8 can be explained based on two opposite effects related to the distribution of light. On the one hand, for the pure photocatalytic process, there is an improvement in photonic efficiency when a greater homogeneous light distribution is achieved as proven above. On the other hand, this effect is counteracted by a higher inactivation of the bacteria when they are subjected to a high UV intensity. This second effect, specific for the inactivation of microorganisms and not applicable to the oxidation of chemicals, makes the disinfection processes relatively independent on the light distribution of the incident radiation.

In terms of energy consumption, Fig. 10 shown that there is a clear improvement in efficiency for the LED-based systems when compared with the Hg-FL. In this case, the differences in photonic efficiency are negligible (Fig. 9), whereas the higher electricity to light conversion efficiency of the LED devices dominates the global energy efficiency of the process. Moreover, the greater energy efficiency of the LED when working at low currents also matches the 40-LED as the most efficient system for bacterial inactivation. This fact would be even more favored if working with higher currents being this system the best option among the studied ones.

4. Conclusions

A careful analysis of LED-based illumination devices for photocatalytic reactors showed that in comparison with traditional mercury lamps they are obviously more efficient in the generation of light, but can lead to a high inhomogeneous radiation field in the reactor. From experiments of photocatalytic oxidation of methanol, it was concluded that an improvement in the distribution of light produces a significant increase in the overall photonic efficiency of the reactor. In contrast, for bacterial inactivation, this higher photonic efficiency is counteracted by the improved bacterial inactivation achieved when the suspensions are subjected to locally high UV intensities.

Both in oxidation of chemicals and in bacterial inactivation it can be concluded that the best option in terms of energy consumption is the option with the highest number of LED due to the improvement of the light homogeneity and energy efficiency when they operate at lower electrical current intensities. Although this conclusion has been derived from a specific tubular reactor configuration, it can be obviously extrapolated to any other geometry and reactor design, suggesting that the illumination device should be carefully designed to get the optimal light distribution for each specific setup.

The main conclusion of this study is that although LED offer numerous advantages over traditional lighting systems, it is critical that the reactor design takes carefully into consideration the light distribution, because otherwise the overall efficiency of the process could be not improved with respect to traditional mercury lamps.

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